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On the possibility of the detection of double quantum emission in atomic nuclei, with special application to the decay of Xe^{131m}

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With 6 figures in the text

ABSTRACT

Some possibilities for the observation of double quantum emission from an excited nuclear level are considered. In particular the decay of the isomeric $h_{11/2}$ level in Xe^{131} has been experimentally studied with emphasis on this second order effect. The measurements have been performed by observation of the coincidence relations between X-rays and gamma- or X-radiation. A number of processes, which can possibly contribute to the results obtained, are discussed in detail. The possibility that double quantum emission has been observed remains then as the most probable one. With this assumption it is possible to estimate an experimental value for the ratio between the transition probabilities for double and single quantum emission in the decay of Xe^{131m} to about one part in a thousand.

Introduction

In the transition between two energy levels in the nucleus, a single quantum[1] usually takes care of the whole energy difference. The transition can, however, also take place by means of second order processes, such as a simultaneous emission of two gamma quanta, two electrons or one gamma quantum and one electron. The probability of this process, however, should usually be several orders of magnitude lower than that of the single quantum emission. The theory of double quantum emission was developed in the thirties by M. Göppert-Mayer [1] and is also treated by R. G. Sachs [2] and more recently by J. Eichler and G. Jacob [3]. This second order process is distinguished from a single quantum transition by the fact that the energy is divided between the two emitted quanta giving rise to a weak continuum. Cf. Fig. 1. No definite experimental observation of the effect seems to have been published up to now. However, the possibility of double quantum emission in the decay of Ir^{192m} has been discussed [15], but the detection seems very doubtful [16]. In the following some details will be discussed on the possibilities for the observa-

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[1] In the following the notation "quantum" represents both photon and electron.

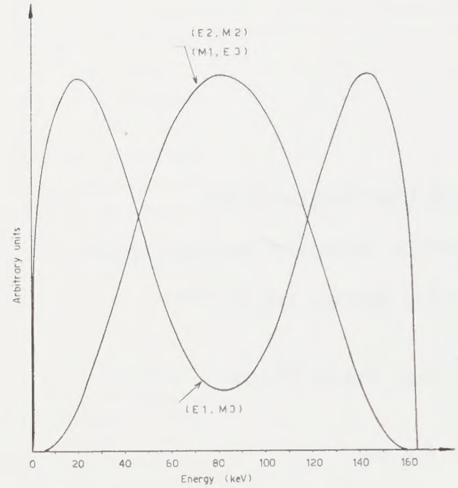

Fig. 1. Theoretically calculated distributions for $E2M2$, $E1M3$ and $E3M1$ transitions according to ref. [3]. The maximum energy chosen is 164 keV ($h_{11/2}$ level in Xe^{131m}). The relative intensities between the distributions are arbitrary.

tion of double quantum emission. The decay of the $h_{11/2}$ level in Xe^{131} will be treated especially, as it has been studied experimentally with emphasis on this second order process. A short note on this subject has recently been published elsewhere by the authors [4].

Possibilities for the detection of double quantum emission

The shape of the distribution of the transition probability, and thus the spectrum of the emitted radiation of double quanta can be very sensitive to the type of transition. As an example, Fig. 1 shows the theoretically calculated gamma ray distributions (according to Eichler and Jacob [3]) for $E2M2$, $E1M3$ and $E3M1$ transitions. These are the possible double quantum transitions competing with an $M4$ single transition. In reality one gets a superposition of the different types of distributions. However, it would, in principle, be possible to compare the matrix elements for the transitions from an experimentally measured distribution.

From an experimental point of view it would be very difficult to observe directly the weak, two quantum distribution as it is usually masked by other processes. When studying the gamma radiation, the Compton process and the bremsstrahlung, for example, will effectively conceal the double quantum effect. It therefore seems natural to investigate this second order process by using some coincidence technique. It would then be possible to select an energy region of the distribution at the energy E for one channel in the coincidence arrangement and to move the other channel over the energy $(E_{\max} - E)$, where $E_{\max}$ is the total transition energy. The existence of double quantum emission will then be proved by a peak on the coincidence counting rate curve at the energy $(E_{\max} - E)$. Due to the relative weakness of the distribution, this method is rather tedious. Another possibility is to study coincidences with the X-rays emitted in connection with the internal conversion of one or both of the gamma quanta. The advantage would be that it is possible to detect transitions of different energies in a narrow region (as X-rays) which makes the observation more favourable compared with a distribution measurement, at least in decays where one has large internal conversion coefficients. A drawback is, of course, the difficulty in interpreting the results due to different channel efficiency for different energies as the internal conversion coefficient changes with the energy and the multipole order of the transition. On the other hand this fact can make it possible to distinguish between cases which have the same shape of the transition probability distribution for gamma rays, as will be seen later.

When trying to detect the double quantum emission one generally needs a nucleus, the decay of which contains only two levels with electromagnetic transitions between them. Then the possibility for detection depends on how favourable the ratio is between the transition probabilities for double and single quantum emission and these are mainly due to the spins and parities of the initial and final energy levels in the nucleus. One thus has to search for decays in which the single quantum transition is hindered. Such cases may be found within the following groups:

A. The $0+\rightarrow 0-$ transition may seem to be the most favourable case as it is strictly forbidden for emission of a single photon or electron or for pair production. The de-excitation can thus take place only through a two quantum process i.e. one $E1$ and one $M1$ transition. No nucleus decaying by such a transition is, however, known to date. Another possibility would be the de-excitation of a $0+$ level to another $0+$ level which goes via an electric monopole transition, where the electromagnetic radiation is strictly forbidden. There may, however, also be an inner pair production, if the amount of energy is large enough, as well as a double quantum emission. This latter effect would thus give rise to all gamma radiation from the de-excitation. It should therefore i.e. be suitable to study the de-excitation of the 1.73 MeV level in Zr^{90} fed by the beta decay of Y^{90}. Experiments have been done along these lines by the authors using the above, first-mentioned coincidence technique. It seems to be possible to detect a very weak effect. The observation is, however, much complicated due to the intense bremsstrahlung connected with the beta decay. The 2.3 MeV ground-state beta transition contributes to more than 99 % of all the decays.

B. Another possible group contains the cases where the single quantum emission is strongly hindered in K-forbidden transitions. It is possible that double quantum emission is not hindered to the same extent and it would then be reasonable to try to detect the effect in this case. However, all known K-forbidden transitions are accompanied by other gamma transitions, making the observation very difficult.

C. A third possibility to detect the double quantum emission occurs in connection with a transition of high multipolarity. The transition rate may then be enhanced by the emission of two quanta of lower multipolarity. The single particle model gives for the ratio between transition probabilities for double and single gamma emission, according to [3] approximately

$$\frac{T(L,L')}{T(L_0)}=\frac{1}{137}\left(\frac{1}{2}\right)^{2L_0+2};\quad (L_0\geqslant 2;\ L+L'=L_0), \tag{1}$$

where L, L' are the multipole orders of the double quantum transitions and L_0 the multipole order of the single quantum transition. It is seen that a low multipole order will give the most favourable ratio. For this reason the 513 keV $M2$ transition in Rb^{85}, for example, seems to be suitable.

However, one can also expect that it would be possible to detect double quantum emission as a competing process to isomeric $M4$ transitions. Thus the 662 keV $M4$ transition in Ba^{137} has been investigated with the coincidence technique. The results

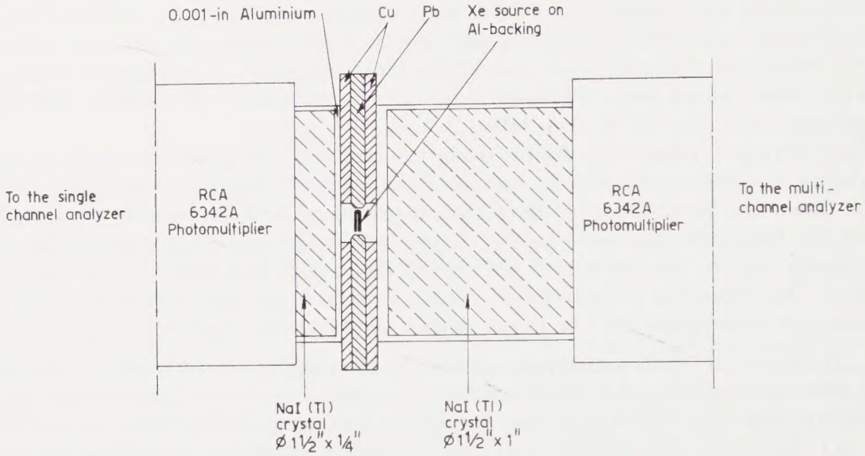
0.001-in Aluminium
Cu
Pb
Xe source on Al-backing
To the single channel analyzer
RCA 6342A Photomultiplier
RCA 6342A Photomultiplier
To the multi-channel analyzer
NaI (Tl) crystal ø 1½" x ¼"
NaI (Tl) crystal ø 1½" x 1"

Fig. 2. The experimental set-up of the detectors.

indicate the presence of coincidences that may arise from double quantum emission, but no definite conclusions have been drawn. The bremsstrahlung from the decay of Cs^{137}, which in itself also may give rise to two or more photons emitted in coincidence, makes it difficult to interpret the data obtained. Many of the difficulties are, however, avoided in the case of the decay of Xe^{131m}. The isomeric $h_{11/2}$ level in Xe^{131} decays to the $d_{3/2}$ ground state by the 164 keV $M4$ transition with a half-life of 12 days. The relative intensity of any other transition in this decay is estimated to be very low. In our attempt to detect the double quantum emission, we therefore studied the radiation from this decay in more detail.

Measurements on the radiation from the decay of Xe^{131m}

1. *Experimental procedure and result*

The source used was prepared by electromagnetic isotope separation [5] of Xe^{131m}. This activity had earlier been milked from I^{131}, which feeds the $h_{11/2}$ level in Xe^{131} by about one percent beta branching. The reason for the use of the relatively complicated isotope separation procedure was two-fold. Firstly, one obtains in this way a small and well defined sample area. Secondly, the possibility of having a radioactive impurity in the Xe-source other than with mass number 131 is negligible, a detail which in the present case is of the utmost importance. Generally it can be stated that one large difficulty in measurements of the type discussed here is just radiation from undesirable and unknown activities present in the source. This can to a large degree be avoided by use of electromagnetically isotope separated samples.

The Xe^{131m} was collected on a 13 mg/cm² thick aluminium backing. The strength of the Xe^{131} sample used in this investigation was about 0.35 μC. The source was mounted between two NaI(Tl) crystals having diameters of $1\frac{1}{2}''$ and lengths of $1''$ and $\frac{1}{4}''$ respectively, both mounted on RCA 6342A photomultipliers with 180° geometry (Fig. 2). The back-scattering from one crystal to the other was hindered

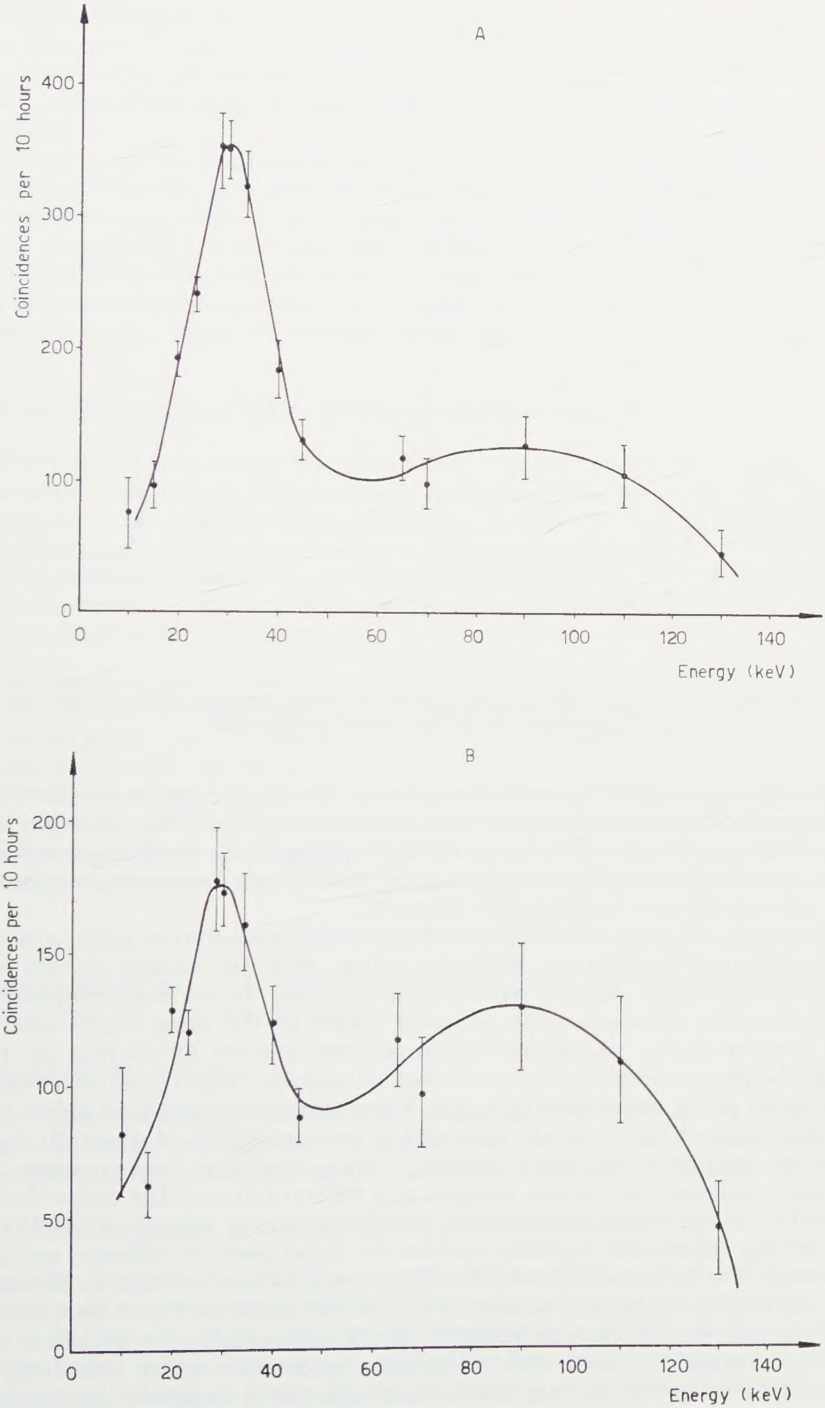

Fig. 3. The coincidence counting rate in the X-ray peak for different positions of the single channel. The peak at about 30 keV is due to coincidences between two X-rays. A) The experimentally obtained points. B) The curve in figure A corrected for double coincidence efficiency in the X-ray region.

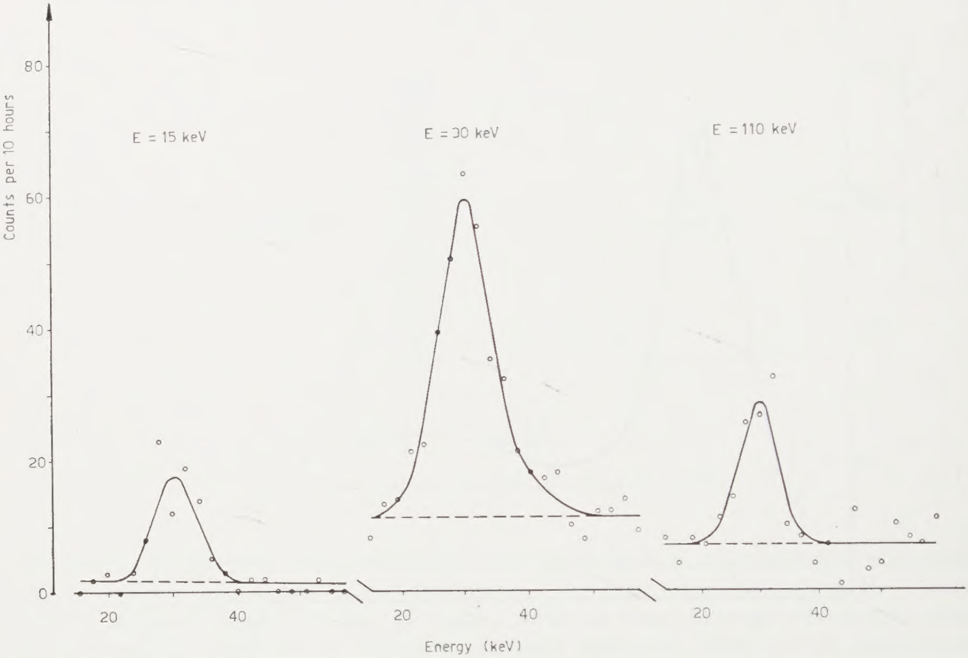

Fig. 4. Spectra obtained in the multi-channel analyzer for some positions of the single channel. The dashed lines are the estimated background.

by a lead and copper shield between the crystals. The source was so mounted that it was surrounded by an aluminium foil and organic material together having a total thickness of 30 mg/cm^2. A careful study of the stopping power for this arrangement shows that the conversion electrons penetrating it can give a maximum contribution of 1 % to the coincidence counting rate obtained.

The pulses from the larger crystal were fed to a multi-channel pulse height analyzer of the Hutchinson-Scarrott type, while the pulses from the smaller crystal were introduced into a single channel pulse height analyzer. In all measurements the single channel with constant width accepted pulses of the same height and the different positions in the gamma ray spectrum were selected by varying the high tension on the photomultiplier. The multi-channel analyzer, which in all the measurements recorded pulses corresponding to the X-ray region, was gated by pulses from a coincidence unit of the slow-fast type with a time-resolution of about 30 mμsec described by Johansson [6]. Since stability during long-time measurements was needed, the coincidence circuit was changed to a differential one. The single channel was allowed to accept pulses corresponding to different energy regions up to 165 keV. Fig. 3 shows the coincidence counting rate for the X-ray peak for different positions of the channel. The background under the X-ray peak, as it is recorded in the multichannel analyzer, is subtracted as seen in Fig. 4. The points in Fig. 3 thus indicate the counting rate for coincidences between pure X-rays and X-rays or gamma rays. It would be difficult to obtain this information quantitatively by measuring the whole spectrum up to 165 keV in coincidence with the X-radiation. Such a coincidence measurement has, however, also been performed showing qualitatively the same feature as in Fig. 3. Corrections due to the decay of the sample are made and the points in Fig. 3B are also corrected for the fact that, when both channels accept pulses arising from gamma rays of the same energy, the coincidence efficiency is doubled.

From Fig. 3 it is seen how the coincidence counting rate has a maximum when the variable channel accepts pulses due to gamma rays with an energy of about 30 keV, that is, the energy of Xe X-rays. Thus, real coincidences between two photons of the X-ray energy have doubtlessly been observed in the experiments. Furthermore, there is a broad distribution of the counting rate for coincidences between X-rays and gamma radiation of energy up to about 130 keV. This energy is about the total transition energy minus the binding energy for the K electron.

2. *Processes other than double quantum emission which can give rise to coincidences*

Before trying to estimate the transition probability for double quantum emission obtainable from Fig. 3, it is necessary to discuss other processes which could also contribute to the coincidence counting rate. It should be remarked that no genuine coincidences with X-rays can occur to a first approximation by measurements on the gamma radiation from the decay of $\mathrm{Xe}^{131\mathrm{m}}$.

1. *Impurities in the* $\mathit{Xe}^{131\mathrm{m}}$ *sample*

This can give rise to true coincidences between gamma rays and X-rays. As the sample was electromagnetically isotope separated, only activities with mass number 131 could occur, i.e. I^{131}. Small amounts of this isotope were actually present. The activity due to this was found to be less than 0.2 % of the Xe activity. The contribution to the observed coincidence counting rate from I^{131} was investigated by use of a pure I^{131} source which was about 40 times stronger than the impurities of I^{131} in the $\mathrm{Xe}^{131\mathrm{m}}$ source. From this measurement it was possible to estimate the coincidence counting rate C_{I} due to the I^{131} admixture at the X-ray peak. One finds

$$C_{\mathrm{I}} \approx 5 \text{ counts per 10 hours.}$$

A test measurement on the distribution showed that the coincidence counting rate there was of the same magnitude as C_{I}.

2. *Accidental coincidences*

The contribution from this cause was measured to be less than 5 % of the observed peak coincidence counting rate by introducing a delay line of about 200 mμsec. A measurement without the source gave negligible coincidence counting rate.

3. *Coincidences between K X-rays and K conversion electrons*

In order to make it possible to measure also gamma rays of low energy in the X-ray crystal, the amount of material between the source and this detector corresponds only to 30 mg/cm², which decreases the intensity of a 10 keV gamma radiation by about a factor of two. However, in spite of the fact that the range of electrons with an energy of 130 keV (K 164) is 20 mg/cm², there is a possibility for some K electrons

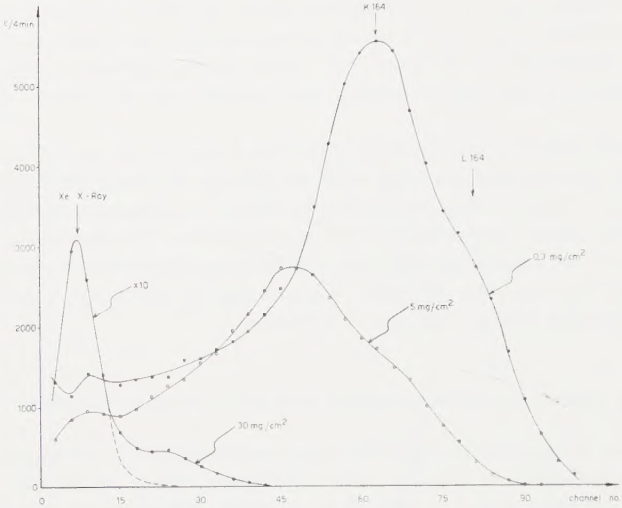

Fig. 5. Electron intensity distributions of the $(K+L)$ 164 keV conversion lines recorded in an anthracene crystal with different absorber thicknesses.

to penetrate into the X-ray crystal. These can therefore give rise to real coincidences. It should be mentioned that the absorber thickness in front of the larger crystal was much more than 30 mg/cm^2.

To investigate the number of K electrons which could be detected by the NaI(Tl) crystal, the electron spectrum from Xe^{131m} was measured by use of an anthracene crystal for different thicknesses of the absorption material. Fig. 5 shows some typical cases. The peak at low energy is due to the X-rays. Even if the resolution is bad, some conclusions can be drawn from this figure. It is seen that the number of electrons which penetrate 30 mg/cm^2 is about 10^{-3} of the initial number. Further, the energy of the maximum of the electron intensity distribution is roughly about 50 keV. The observed intensity seen in Fig. 5, for 30 mg/cm^2 absorber thickness, is probably mainly due to electrons from the L conversion of the 164 keV gamma ray. An upper limit of the number of K electrons penetrating the shield can thus be taken to be 10^{-3}. Further, as the efficiency of NaI(Tl) for electrons is only about 10 %, one can estimate that the total amount of possible X-ray-K electron coincidences must be less than 1 % of the observed peak intensity in Fig. 3. As all coincidence measurements have been performed by use of small energy intervals in the channels, it is obvious that coincidences between K-electrons and their corresponding K X-ray is quite negligible.

4. *Multiple scattering between the two detectors*

Backscattering from one crystal to the other, especially of the iodine escape X-ray, can contribute to some extent to the coincidence counting rate. The effects are, however, largely suppressed by use of the lead and copper absorbers between the crystals. To test the effectivity of this arrangement, special measurements were performed with an Hg^{203} sample replacing the Xe^{131} source. Hg^{203} decays exclusively by beta emission to the 279 keV level in Tl^{203}, which decays further by an $M1$ transition to the ground state. It is quite reasonable to assume that the double quantum emission in this transition is negligible. In these experiments no such effect as in Xe^{131m} was observed.

5. *Internal Compton effect (ICE)*

This is a nuclear transition in which there is the simultaneous ejection of a bound electron and the emission of a gamma ray. This effect has been experimentally observed recently [7]. In the present case, possible coincidences can be measured between K X-rays and gamma rays due to the ICE. This would give rise to a continuous distribution (in Fig. 3), the intensity of which would increase with decreasing energy of the gamma ray, as the intensity of the scattered gamma ray increases for lower energies. The cross-section for the process has been calculated by several authors [8], [9], [10]. In the non-relativistic region, neglecting angular correlation and multipole order, one obtains the expression for the differential ICE coefficient per unit solid angle [10]

$$B(q_1 q_2) = \frac{\alpha}{3\pi^2} W \int_{q_1}^{q_2} \frac{dq}{q}, \tag{2}$$

where α = the fine structure constant,
W = energy given up by the nucleus in m_0c^2 units,
q_1 and q_2 = lower and upper limit of scattered photon energy.

The expression (2) has been shown to give the correct order of magnitude of the effect, even if it is somewhat too large in comparison with the more carefully calculated cross-section and the values obtained experimentally in the special case of Ba^{137} [7].

If in the present case we choose a certain energy interval, it is possible to obtain an upper limit of the ICE. As an example, for $q_1 = 80$ keV and $q_2 = 95$ keV, one finds from eq. (2), with $W = 0.32$,

$$B = 1.4 \cdot 10^{-5}$$

When the internal conversion coefficient α is large, as in the present case, the coincidence counting rate is given by the expression

$$C_{\text{ICE}} = N_0\, \omega_1\, \varepsilon_1\, \omega_2\, \varepsilon_2\, \varepsilon_c\, B\, 4\pi \frac{\alpha_K}{\alpha} w, \tag{3}$$

where C_{ICE} = coincidence rate due to ICE,
N_0 = total disintegration rate,
ω_1 = solid angle of the detector of the multi-channel analyzer,
ε_1 = efficiency of the detector of the multi-channel analyzer,
ω_2 = solid angle of the detector of the single channel analyzer,
ε_2 = efficiency of the detector of the single channel analyzer,
ε_c = coincidence efficiency,
w = fluorescence yield.

With the values of N_0, ω and ε given in section 3 A, one finds from eq. (3)

$$C_{\text{ICE}} = 35 \text{ counts per 10 hours.}$$

Even if this is an upper limit, the contribution of the ICE to the coincidence counting rate can be large. It cannot, however, give rise to a peak. It should be observed that in Fig. 3 the contribution of the ICE is partly subtracted (cf. Fig. 4).

6. *External bremsstrahlung from the K conversion electrons*

One may obtain coincidences between K X-rays and photons emitted as bremsstrahlung, the intensity of which should increase with decreasing energy. It should be noted that bremsstrahlung cannot account for the peak in Fig. 3.

However, it may be quite reasonable to assume that the continuous distribution in Fig. 3 could indicate that the bremsstrahlung is of minor importance, as the measured intensity does not increase towards lower energies. This has also been experimentally demonstrated by a test measurement with an element with a high Z value mounted close to the activity. A 5 mg/cm^2 thick Au foil was used. The coincidence counting rate increased then by about 20 %. Due to the large increase in Z, from 13 to 79, in this test, the small increase in the coincidence counting rate indicates that bremsstrahlung is of minor importance.

7. *K electron ionisation of Xe by the emitted K conversion electrons*

This can give rise to real coincidences between two X-rays and must therefore be handled carefully. The amount of stable Xe in the sample was about 1 μg. This comes from the isotope separation procedure where stable Xe gas was used as carrier. To measure a possible K ionisation effect, a large quantity of Te (10 mg/cm^2) was inserted in contact with the sample. As the intensity and mean energy loss of the K conversion electrons in a 1 mg/cm^2 layer is almost negligible (cf. Fig. 5), it is possible to state that one should get a strong magnification (roughly, at least 1000) of the effect in this test experiment. The coincidence counting rate was in fact increased by a factor of two, part of which may be due to increased bremsstrahlung. By comparing this with the expected effect one can conclude that the K ionisation must be completely negligible.

8. *External Compton effect*

This should give a contribution to the continuous distribution. However, according to the measurement discussed in section 7, the result of which also can be applied here, the external Compton effect must be extremely small.

9. *An intermediate level*

This can be a very important competing effect, as one here also obtains real coincidences between two X-rays. Experimentally it has been found that the intensity of a possible conversion line other than the K 164 keV line in the decay of Xe^{131m} must be less than $5 \cdot 10^{-4}$ of the intensity of the K 164 keV line [11]. This information shows definitely that a transition to an intermediate level cannot explain the whole effect in Fig. 3.

From the knowledge of the decay of I^{131} it can be concluded that a single particle level, the $s_{1/2}$ level, can be a possible intermediate state. The decay to the ground state should then go via an $E5$ transition from $h_{11/2}$ to $s_{1/2}$. The transition probability for this is, however, very small, both theoretically (cf. e.g. [12]) and experimentally. The authors have, for example, determined the transition probability for $E5$ transitions in Te^{121m} and Te^{123m} to be less than 10^{-4} of that for the corresponding $M4$ transitions. In the Te cases the energy of the $E5$ transition should be larger than that of the $M4$ gamma ray. In the present case, in Xe^{131m}, however, the energy of $E5$ gamma ray is less than that of the $M4$ gamma ray. This makes the decay $h_{11/2} \rightarrow s_{1/2} \rightarrow d_{3/2}$ very improbable.

Using the intensity limit given above of $5 \cdot 10^{-4}$ for any possible electron line and taking into account that $K/(L+M)$ for an $E5$ transition of energy about 80 keV is as small as 0.009, one can calculate the coincidence counting rate between two K X-rays for such a mode of decay to be only 0.01 counts per 10 hours. There can, of course, exist other intermediate states, but these seem very improbable from a systematic point of view. A careful experimental investigation using a β-spectrometer will, however, be made soon with special attention to this question. In principle, one should also be able to detect an intermediate level as a peak in the continuous distribution. A special search for such a peak has not been performed yet, but its existence seems very unlikely due the reasons given above.

Lastly, it should also be mentioned in this section that the $h_{11/2}$ level in Xe^{131} cannot decay to levels in any neighbouring element, as the ground state of I^{131} is 815 keV above the $h_{11/2}$ level and the ground state of Cs^{131} has an energy of about 170 keV above this level [13].

Among the effects mentioned above, only those discussed in the sections 7 and 9 are able to give rise to the observed peak in Fig. 3. As the effect in section 7 is negligible and that in section 9 is very improbable, the two quantum emission is the only possibility left. This is also supported by the measured distribution is Fig. 3.

3. *Estimation of the transition probability for two quantum emission in* Xe^{131m}

From the discussion in the last section it is seen that the effects competing with two quantum emission as measured in Fig. 3 are of minor importance. They can, in fact, be estimated to be less than 25 % in the distribution while they should be negligible for the peak. To make an estimation of the transition probability possible, we assume therefore that the whole observed effect at the peak is due to double quantum emission.

A. *The observed peak due to X-ray coincidences.*—In order to calculate the double quantum effect from the observed coincidence counting rate, it is necessary to use some kind of mean value of the conversion coefficient, as one has a continuous distribution of the energy of the gamma rays and conversion electrons. In the present case the calculation has been made by dividing the energy region into 10 equal parts and taking the conversion coefficient for the mean energy of the gamma rays in each interval. It should be observed here that the only conversion coefficients tabulated are those which take into account only single electron emission. In the present case, however, two K conversion electrons are ejected simultaneously. For this reason the conversion coefficients are probably not applicable without corrections. It is further assumed that one has an $E2M2$ transition, which gives the maximum intensity at the half value of the maximum energy. Fig. 6 A shows the $E2M2$ distribution divided into 10 parts, each having an energy width of $\Delta E = 1/10 \; E_{\max}$. The energy in the interval n is $E_n = \Delta E(n - \frac{1}{2})$.

The coincidence counting rate C_p due to coincidences between two X-rays is then

$$C_p = \Sigma_n C_p \, (E2M2), \tag{4}$$

where

$$_nC_p \, (E2M2) = 2 \, N_0 \, P_p \, \omega_1 \, \varepsilon_1 \, \omega_2 \, \varepsilon_2 \, \varepsilon_c \, \delta_n \, \frac{_n\alpha_K}{1 + {}_n\alpha} \cdot \frac{_m\alpha_K}{1 + {}_m\alpha} \, w^2, \tag{5}$$

and

$_nC_p(E2M2)$ = coincidence counting rate due to X-rays corresponding to the K conversion electrons. The $E2$ transition has an energy (E_n) corresponding to the interval n. The $M2$ gamma ray has therefore an energy equal to ($E_{\max} - E_n$). It should be remarked that the factor two must be introduced because, when both channels accept gamma rays of the same energy, the coincidence counting rate increases by a factor of two (cf. Fig. 3A and B).

N_0 = total disintegration rate,
P_p = coefficient of double quantum emission, i.e. the quotient between the disintegration rates due to double and single quantum emission, as estimated from the peak,
ω_1 = solid angle of the detector of the multi-channel analyzer,
ε_1 = efficiency of the detector of the multi-channel analyzer,
ω_2 = solid angle of the detector of the single channel analyzer,
ε_2 = efficiency of the detector of the single channel analyzer,
ε_c = coincidence efficiency, equal to one in the present case,
δ_n = the fraction of the distribution corresponding to the interval n,
$_n\alpha$; $_n\alpha_K$ = total- and K conversion coefficient of the gamma ray with energy E_n in the interval n,
$_m\alpha$; $_m\alpha_K$ = total- and K conversion coefficient of the gamma ray with energy ($E_{\max} - E_n$) in the interval $m = 11 - n$,
w = fluorescence yield.

The constants ω_1, ω_2, ε_1, ε_2 have been determined from a coincidence measurement between two X-rays with a $\mathrm{Te}^{125\,\mathrm{m}}$ sample, which has been prepared by electromagnetic isotope separation. It was found that $\omega_1 \, \varepsilon_1 = 0.04$ and $\omega_2 \, \varepsilon_2 = 0.02$. The small value of $\omega_2 \, \varepsilon_2$ is to some extent due to the fact that only a part of the X-ray peak was accepted by the single channel analyzer. The low value of $\omega_1 \varepsilon_1$ is partly due to absorption in the material surrounding the crystal connected to the multi-channel analyzer.

From Fig. 3A one obtains the coincidence counting rate due to two pure X-rays (the estimated background counting rate has been subtracted from the maximum counting rate at the peak)

$$C_p = 200 \pm 35 \text{ counts per 10 hours}$$

It should be observed that Fig. 3B shows the measured counting rate after the corrections for the double coincidence efficiency have been introduced.

Table 1. The values of P (the probability for double quantum emission) obtained for different types of transitions.

Transition type	P_p from eq. (4) by using		P_d from eq. (8) $(n=6)$	$P_d/P_p(\frac{1}{2}\alpha_K)$
	α_K	$\frac{1}{2}\alpha_K$		
$E2\,M2$	$1.7\cdot10^{-3}$	$3.2\cdot10^{-3}$	$1.3\cdot10^{-2}$	4.1
$E3\,M1$	$3.2\cdot10^{-3}$	$5.7\cdot10^{-3}$	$3.9\cdot10^{-2}$	6.8
$E1\,M3$	$3.5\cdot10^{-3}$	$7.6\cdot10^{-3}$	$5.2\cdot10^{-2}$	6.8

To estimate P_p it is necessary to determine what kind of conversion coefficient should be used. In the absence of something more realistic, the values of α_K used are those tabulated [14] divided by two. This is done because in the calculation of α_K it is assumed that one has two conversion electrons for each transition, but in the present case only one K electron is available.

From Fig. 6A and the conversion coefficients treated in this way one finds for an $E2M2$ transition

$$\sum \delta_n \frac{{}_n\alpha_K}{1+{}_n\alpha}\cdot\frac{{}_m\alpha_K}{1+{}_m\alpha}=0.14. \tag{6}$$

Thus, from eq. (4)

$$P_p=3.2\cdot10^{-3}. \tag{7}$$

Table 1 gives the values of P_p for the other possible transitions, $E1M3$ and $E3M1$, obtained in the same way as that for $E2M2$. Further, the values of P_p are also given for the case where no corrections of the tabulated α_K are made.

B. *The continuous distribution.*—From the distribution in Fig. 3 one can also estimate the ratio P_d between double and single quantum emission. Coincidences due to double quantum emission are then measured between gamma rays and K conversion electrons detected by means of the corresponding X-rays. When the single channel accepts a gamma ray of the energy E_n in interval n (see Fig. 6A) and the distribution is of the $E2M2$ type, the coincidence counting rate ${}_nC_d$ is

$${}_nC_d=N_0\,P_d\,\omega_1\,\varepsilon_1\,\omega_2\,\varepsilon_2\,\varepsilon_c\,\delta_n\,K_n\,w, \tag{8}$$

where

$$K_n=\frac{1}{2}\left[\left(\frac{1}{1+{}_n\alpha}\right)_{E2}\left(\frac{{}_m\alpha_K}{1+{}_m\alpha}\right)_{M2}+\left(\frac{1}{1+{}_n\alpha}\right)_{M2}\left(\frac{{}_m\alpha_K}{1+{}_m\alpha}\right)_{E2}\right]. \tag{9}$$

The symbols are similar to those used in eq. (5).

The distribution one should detect when measuring coincidences between X- and gamma rays and taking into account the conversion, is given in Fig. 6B. The value of $\delta_n\cdot K_n$ is there drawn as a function of the energy interval number n for the three competing possibilities, namely $E2M2$, $E1M3$ and $E3M1$. It is seen that the distribution for an $E2M2$ transition is not longer symmetrical, but the maximum is shifted towards higher energy. For the type $E3M1$ the asymmetry is not so pronounced. It should be noted here that a transition $E3M1$ seems to be the most probable one in the case of $\mathrm{Xe}^{131\mathrm{m}}$, as the nearest known level above the $11/2^-$ state is a $5/2^+$ state. A possible second order transition may then be $11/2^-$; $5/2^+$; $3/2^+$ which could

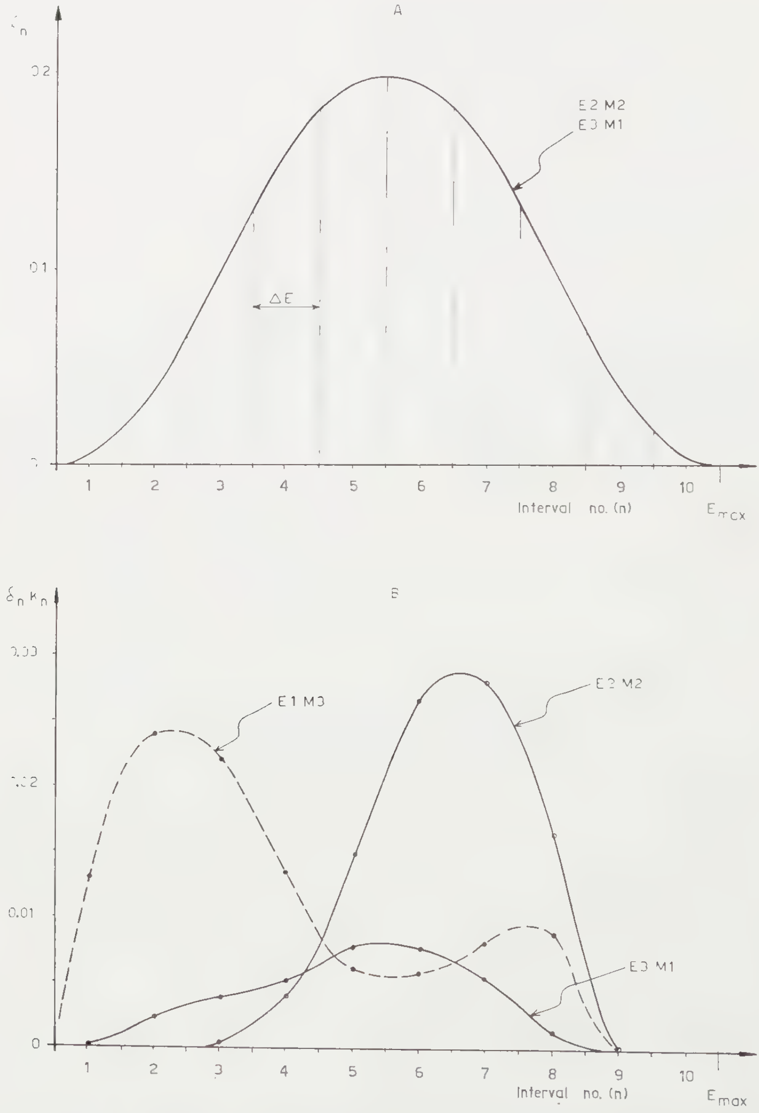

Fig. 6. A) shows δ_n (the fraction of the distribution corresponding to the interval n) for different values of the 10 energy intervals used. The disbribution is of the type $E2M2$ and $E3M1$ (cf. Fig. 1). B) shows $\delta_n K_n$ (cf. eq. (8) in text) for the different energy intervals for the three possible types of transition, viz. $E2M2$, $E3M1$ and $E1M3$. These curves therefore give the distributions one should find when measuring coincidences between gamma- and X-rays with the constant channel width ΔE in figure A. It should be noted, however, that these curves therefore need a small correction to be comparable with the experiments, as in these a constant relative channel width was used.

give just an $E3M1$ type transition. However, levels occur also with higher energies, having other spins and parities. Other types of transitions may then take place via these states.

Proceeding in the same way as in the last section in calculating the coincidence counting rate and assuming an $E2M2$ transition, one finds from eqs. (8) and (9)

$$P_d = 1.3 \cdot 10^{-2}. \tag{10}$$

This value is obtained for an energy of the gamma ray of about 90 keV (channel number 6 in Fig. 6A; the experimental channel width was just 16 keV for the 90 keV position). The coincidence counting rate used is 90 counts per 10 hours, which is found from Fig. 3 after a correction of 25 % which may be due to processes other than double quantum emission (cf. section 2). Table 1 gives the values of P_d for the two other possible distributions, $E1M3$ and $E3M1$.

Owing to the large uncertainty of the observed distribution and the difficulties in distinguishing this type from any other, this calculated value of P_d must be handled very carefully. The error due to conversion coefficient is not present in P_d, as in this case only one conversion electron is ejected. The tabulated conversion coefficients can therefore be used. The value of P_d estimated in this way should thus be larger than the value of P_p. The hindrance factor due to simultaneous emission of two K conversion electrons could therefore be estimated. In the present case one should have for an $E2M2$ transition, for exemple, a hindrance factor, P_d/P_p, of about 4.

4. *Comparison between experiment and theory*

The value of the ratio of the transition probability for double to single gamma emission in $\mathrm{Xe}^{131\mathrm{m}}$ as calculated by Eichler and Jacob [3] is $5 \cdot 10^{-5}$. It should be remarked that this value is somewhat larger than that the approximate relation (1) gives, which is $7 \cdot 10^{-6}$ for the present case. The nuclear potential used by Eichler and Jacob is of an oscillator type without spin-orbit coupling. It is difficult to estimate which of the two processes, single or double quantum emission, should be favoured by applying a more complete nuclear potential. The experimental data, as found in this work, seem to show that the second order process is more common than the calculated ratio given above would indicate.

To compare the observed ratio P, i.e. the number of double quanta to single quantum with the calculated transition ratios, one must make some very uncertain assumptions, especially concerning the conversion. If λ is the total decay constant of the $h_{11/2}$ level in $\mathrm{Xe}^{131\mathrm{m}}$ and λ_γ and λ_e the decay constants for the emission of only gamma rays and only conversion electrons, respectively, one has, if λ_2 is the decay constant due to the two quantum emission and λ' represents other higher order processes,

$$\lambda = \lambda_\gamma + \lambda_e + \lambda_2 + \lambda'. \tag{11}$$

λ_2 can be written

$$\lambda_2 = \lambda_{\gamma\gamma} + \lambda_{e\gamma} + \lambda_{ee}, \tag{12}$$

where

$\lambda_{\gamma\gamma}$ = decay constant for gamma-gamma emission,
$\lambda_{e\gamma}$ = decay constant for electron-gamma emission,
λ_{ee} = decay constant for electron-electron emission.

From (11) and (12) one finds if neglecting λ_2 and λ' in eq. (11)

$$\frac{\lambda_2}{\lambda}=\frac{1}{1+\alpha}\cdot\frac{\lambda_{\gamma\gamma}}{\lambda_\gamma}\left(1+\frac{\lambda_{e\gamma}}{\lambda_{\gamma\gamma}}+\frac{\lambda_{ee}}{\lambda_{\gamma\gamma}}\right). \tag{13}$$

Here λ_2/λ is the measured ratio P and $\lambda_{\gamma\gamma}/\lambda_\gamma$ is the ratio which should be compared to the theoretically calculated value. α is λ_e/λ_γ.

If it is possible to write, in analogy with the relation $\lambda_e=\alpha\lambda_\gamma$,

$$\lambda_{e\gamma}=A_{e\gamma}(EL\,ML')\,\lambda_{\gamma\gamma}, \tag{14}$$

$$\lambda_{ee}=A_{ee}(EL\,ML')\,\lambda_{\gamma\gamma}, \tag{15}$$

where

$$A_{e\gamma}(EL\,ML')=\frac{1}{2}\sum\delta_n\,[{}_n\alpha(EL)+{}_m\alpha(ML')], \tag{16}$$

$$A_{ee}(EL\,ML')=\frac{1}{2}\sum\delta_n\,[{}_n\alpha(EL){}_m\alpha(ML')+{}_n\alpha(ML'){}_m\alpha(EL)], \tag{17}$$

then one can get an estimation of $\lambda_{\gamma\gamma}/\lambda_\gamma$ from eq. (13). The symbols in eqs. (14)–(17) are the same as those used above. Cf eq. (5). Table 2 shows the results of such a calculation, if the value of P in eq. (13) is equal to P_p ($\frac{1}{2}\,\alpha_K$) given in Table 1.

Table 2. The experimental value of $\lambda_{\gamma\gamma}/\lambda_\gamma$ as obtained from the measured value of P_p.

Type of transition	$\lambda_{\gamma\gamma}/\lambda_\gamma$
$E2\,M2$	$8\cdot10^{-4}$
$E3\,M1$	$1\cdot10^{-4}$
$E1\,M3$	$1\cdot10^{-5}$

The measured distribution (Fig. 3) is very uncertain, but it may indicate that the type of transition in the present case probably is $E2M2$ or $E3M1$ or both of them. Cf. Fig. 6B. The experimental value of $\lambda_{\gamma\gamma}/\lambda_\gamma$ seems therefore to be larger than the calculated ratio, roughly by one order of magnitude. The same conclusion can be drawn from the value of P_d but this is still more uncertain.

Conclusion

From the discussion given above it should be clear that the possibility to observe double quantum emission from nuclear excited states is not quite excluded. The measurements made on Xe^{131m} seem to indicate that the second order effect can in some favourable cases be an order of magnitude larger than what calculations on the transition probability seem to indicate. As the double quantum emission should be increased when the single quantum transition is of lower multipolarities than in the case of Xe, a search for the second order process in, for example, a transition of the

$M2$ type seems very interesting. Thus, if the measurement in the present investigation indicates a real effect, one should find the significance in the $M2$ case an order of magnitude larger.

It is necessary, of course, to be extremely careful in attempting an interpretation of the reported data. With the reservation that any at present unknown effect perhaps could explain the results of the measurements in another way, it seems to us very probable that double quantum emission has here been experimentally verified. The discrepancy between the experimental results and the theoretically calculated values is perhaps not astonishing considering the theoretical difficulties as well as experimental uncertainties.

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